Radiolysis Process for the Regeneration of Sodium Borate to Sodium Borohydride

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Objectives:

Develop a viable method for regenerating sodium borohydride from sodium borate, to meet DOE's fuel cost target of \$1.50/kg H_2.

- Demonstrate radiological methods of converting borate to borohydride
- Validate earlier observations, outcomes, and results
- Initiate processes for identifying, qualifying and quantifying conversion mechanisms
- Estimate production capability of process

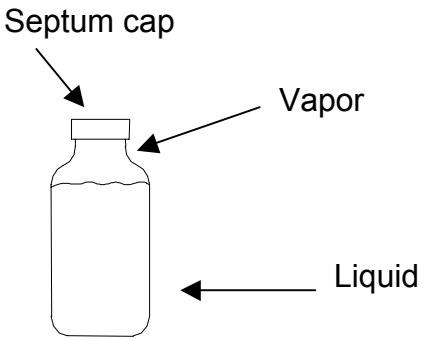
Targets and Barriers

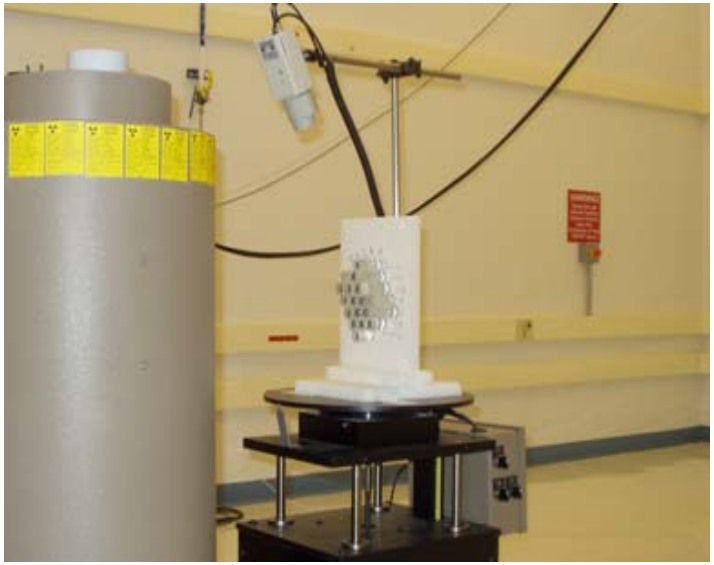
- DOE Technical Barriers for Chemical Hydride Storage Systems
  - A Cost
  - C Efficiency
  - G Life Cycle and Efficiency Analysis
  - Q Regeneration Processes for Irreversible Systems
  - R By-Product Removal

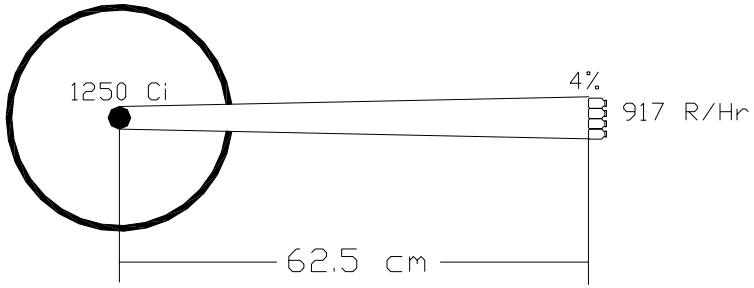
- DOE Technical Targets for Chemical Hydride Storage Systems for 2010
  - Fuel Cost $1.50 per gasoline gallon equivalent

Procedure

- Radioactive sources used (Cs^{137} ,Co^{60} ,Sr^{90} ,X-ray)
- Both Tetraborate and Metaborate tested at controlled concentrations
  - Tetraborate $Na_2B_4O_7$
  - Metaborate $NaBO_2$
- DI water sample used for baseline comparison
- Vapor space sampling of converted hydrogen
- NMR and XRD analysis
- 1,3,4,7,14, and 20 day samples tested thus far







Challenges

- Very little information is known about borate radiochemistry mechanisms and reactions
- Limiting borate solubility's in water
- Limiting detection methods for discriminating converted borohydride (analytical issues)
- Aqueous stabilization issues of borohydride (prevent back reaction to borate)

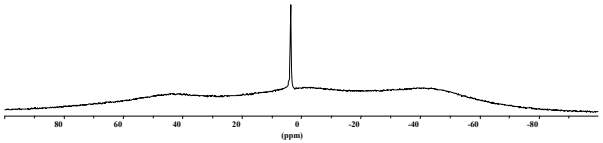
Project Safety

- Safety Analysis Processes
  - Independent Hazard Review Process
  - Environmental Checklist
- Hazards Identified
  - Chemical hazards (mitigate by using proper PPE)
  - Hydrogen gas and air mixtures
  - Pressure building in bottles due to the release of Hydrogen and Oxygen (mitigate by controlling sample size)
  - Radiation exposure (technical, facility, and security processes to minimize radiation exposure)

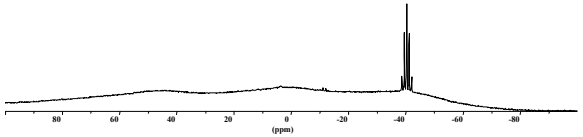
Technical Accomplishments

- Duplicated FY02 tests in Jan and Mar/Apr 2004 (5 times equivalent hydrogen production quantified)
- Duplicated the ability to generate hydrogen from borate solution (3 times in CY04 obtained the same yield efficiency)
- Developing analytical method/process
- Quantified absorbed radioactive energy
- Have some indication analytically of the possibility of borate to borohydride generation
- Vary parameters to increase ability to detect borohydride (e.g., nmr, x-ray diffraction)
  - Use solid or paste borate material
  - Use more radioactive energy per time
  - Work to provide stabilized aqueous solution for borohydride
  - Dehydrate borate/borohydride solution
- Radiolysis of borate showed 53% conversion efficiency

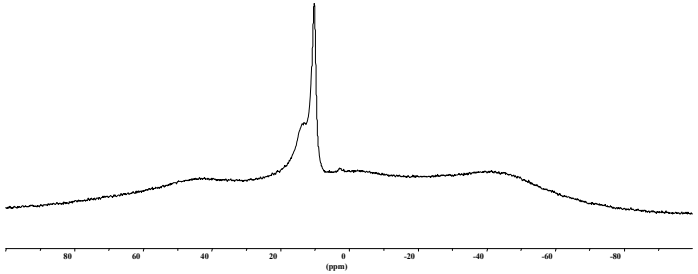
NMR Spectra of Control and Actual Samples



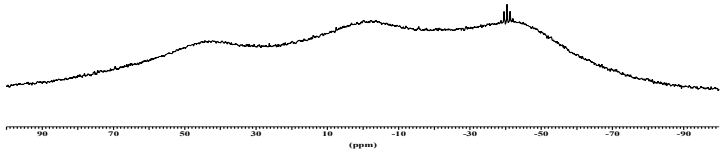
BNMR (sodium borate - control sample)



BNMR (sodium borohydride – control sample)

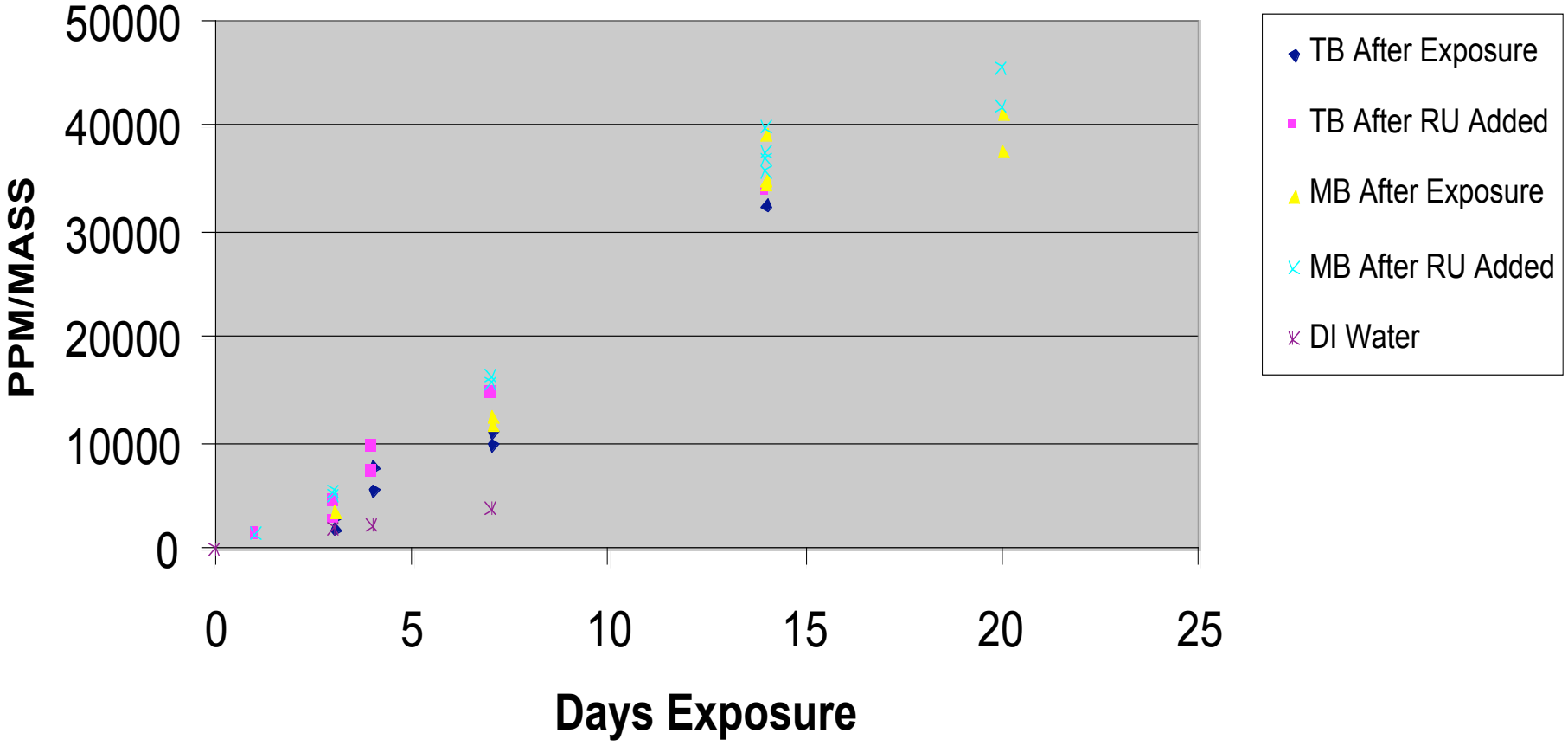


^{11}B NMR (sodium borate peak)



^{11}B NMR (sodium borohydride peak)

Hydrogen Production from Borate Conversion



Radioactive Waste Energy Rough Estimates

Commercial Reactor Waste

- Commercial power plants produce 7.52E9 Ci/yr-reactor waste Approximately 103 reactors in the U.S. that have been running for 25 years.
- Spent waste is being removed from the reactors every 18 months
- Assuming new waste replaces waste with low decay rates and using an average 2.28E8 Ci/yr-reactor.
  - Borate to borohydride conversion requires 1440 kJ/mole
  - 325 M kg/yr Hydrogen produced per year
  - At $1.50/kg Hydrogen production creates $486 M per year
  - Reclassifies waste into a usable product

Commercial Reactor Radiation

- Utilizing 10% of the available radiation in a commercial reactor
- 1.0 B kg/yr of Hydrogen produced per reactor

Interactions and Collaborations

- Millennium Cell
  - Discussed radiation concepts and conversion rates
  - Collaboration on the use of ruthenium
- Idaho State University
  - Radiation measurement to verify amount absorbed
  - Analysis of chemical reactions

Technical Team's Comments and Resolutions

- Verify identification of borohydride (e.g., nmr, x-ray diffraction)
- Verify 53% conversion
- Determine mass and energy balances
- Investigate methods to eliminate or greatly reduce aqueous back reaction of borohydride
- Continue to provide more data

Future Work

- Continue investigating methods of improving efficiency and yield
- Identify methods to control back reactions of borohydride
- Quantify the impact of different radiation sources
- Investigate the impact of catalysts
- Qualify conversion mechanism
- Applicability of this process to other chemistries

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